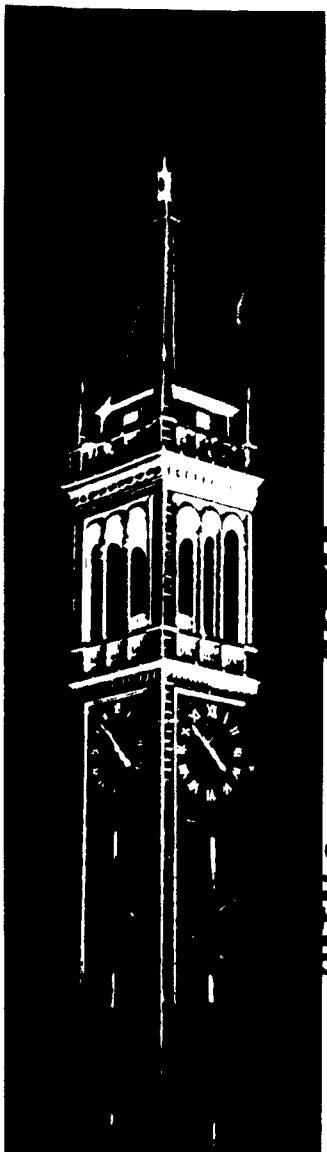

Photodissociation of Molecules For Excitation of an Optical Maser

by

J. R. Singer

Series No. 60, Issue No. 429
Contract No. AF 49(638)-102
January 8, 1962

ELECTRONICS RESEARCH LABORATORY

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One of the most interesting aspects of optical masers is the fact that a very narrow bandwidth wave is obtainable in the infrared and visible wavelengths. The bandwidth of the output signal $\Delta\nu$ is related to the molecular bandwidth $\Delta\nu_a$ by the formula[1]

$$\Delta\nu = \frac{4\pi kT(\Delta\nu_a)^2}{P_a} \tag{1}$$

where k is Boltzmann's constant, T is the absolute temperature, and P_a is the power emitted by the atomic transitions. It is advantageous to obtain a very narrow emission source for a number of reasons; perhaps the major reason is that the peak intensity of the source is enhanced, and the coherence is also usually increased. Intense coherent sources are of interest to provide a new means of studying molecular bonding, harmonic excitation of molecular vibrations, and they may be utilized as communication channel carriers in addition to a large number of other possibilities.

The best method of obtaining a very narrow optical emission source is to eliminate doppler broadening by observing atomic transitions perpendicular to the path of an atomic beam. This method can be adapted to optical masers and we will discuss our approach to this problem.

The system which we are presently investigating involves the photodissociation of molecules in which one dissociated atom appears in an excited state. The general process may be described by

$$(1)\quad h\nu_o + AB \rightarrow A^* + B' + K.E.$$

$$(2)\quad A^* \rightarrow h\nu_1 + A$$

where $h\nu_o$ is a high energy photon interacting with a molecular compound AB composed of atoms A and B. The dissociation of AB leaves A in an excited state and B in the ground state or in a metastable state. KE is the kinetic energy available to conserve energy in the reaction. Following the dissociation, A^* decays to the ground state with the emission of a photon $h\nu_1$ which is lower in energy than $h\nu_o$.

There is the advantage here that the excitation occurs by a band absorption, thus an incoherent radiation source may be used efficiently. On the other hand, the dissociative process has a threshold, and the

exciting light frequencies exceeding the threshold provide energy which reappears as kinetic energy of the dissociated atoms. This kinetic energy is not desirable because it increases doppler broadening of the emission.

If a static system of molecular vapor is dissociated by radiation, the process is necessarily pulsed with a very long recovery time since the dissociated atoms A and B will not, in general, recombine rapidly. The most appropriate system, therefore, is a molecular beam configuration.

If the lifetime of the excited atomic state occuring after dissociation is sufficiently long, the optical maser system configuration may consist of a molecular beam passing through an illuminated region where the molecule is dissociated; the excited atoms then pass into a region surrounded by a Fabry-Perot etalon (or a confocal spherical optical resonator) where coherent radiation could take place. This system required atomic lifetimes of at least 10^{-5} seconds since molecular beam velocities are of the order of 10^5 cm/sec. It is, however, more usual to observe lifetimes of 10^{-8} seconds.

With the shorter atomic lifetimes, the excitation by dissociation must take place within the optically resonant structure. Also, shorter lifetimes imply more difficulty in obtaining continuous operation since the ground state atoms may accumulate faster than they can be swept out of the active region of the maser. That is, the molecular beam velocity may be too slow relative to the radiative lifetime to permit the inverted population distribution to exist for an extended time. The factors leading to a restoration of the Boltzmann distribution are spontaneous emission, and the expectation that atoms attaining the ground state will not be re-excited by the UV pumping radiation, but rather will be acting as absorbers of the coherent output.

Some molecules which are candidates for a molecular beam optical maser are listed in Table 1.

While it would be desirable to select a transition with a long lifetime because then excited atoms could be accumulated, however, the transitions listed here are strong transitions with consequent short-lived states. The substance which we are presently investigating is RbI. The advantages of this material is that the excitation source is easily obtained in the form of the 2537 Å line of a mercury lamp, and that the emission is conveniently in the near infrared.

The proposed experimental setup is shown in Fig. 1. The general plan is to photodissociate the molecules within an optically resonant structure and have the excited atoms decay with the emission of coherent light because of the positive regeneration of the structure.

As mentioned previously, it is desirable to have a very narrow atomic linewidth. The major broadening mechanism is the doppler shift, hence one wants a molecular effusion process from the oven rather than turbulent flow. The requirement for a molecular effusion is from a thin walled aperature[3]

$$\omega \sim L$$

where ω is the slit width, and L is the mean free path within the oven. Assuming 10. mm of Hg pressure within the oven, (. 03 cm for the oven mean free path), the number of molecules effusing per second is given by

$$I \approx 10^{22}\, p\, A_s / (MT)^{1/2}$$

where p is the pressure in mm of Hg, A_s is the area of the source, M is the molecular weight, and T is the absolute temperature (approximately 1000°K for RbI). Using a meter long oven, one obtains a beam intensity of approximately 10^{21} molecules/second. While this beam is not well collimated it enters the emissive region essentially without loss of molecules because of the closeness and breadth of the optically resonant region.

Assuming reflecting areas of 4 cm^2 separated by a meter for the optically resonant region, the number of molecules from the beam within the region is about 10^{16} molecules.

We now ask is this sufficient to satisfy the oscillation condition, [4]

$$N_{min} \geq \frac{3hV\Delta\nu}{4\pi^2\mu^2 Q}$$

and observe that N_{min} is about 10^{14} to 10^{15} molecules, hence the oscillation condition should be satisfied providing the cross-section for the photodissociation is sufficiently large. Sufficient leeway is available so that dissociation need not occur in all of the molecules, but we would hope to dissociate at least ten percent of the molecules within the resonant region. Previous work has demonstrated that photodissociation of RbI is easily observed, [5, 6] but little quantitative data on cross-sections is available.

TABLE 1

Molecule	Metal atom emission λ output	Minimum excitation λ	Metal Ion Emission Transition
LiI	6708A	2350A	$2^2P_{1/2}$--$2^2S_{1/2}$
NaI	5896	2450	$3^2P_{1/2}$--$3^2S_{1/2}$
NaI	3303	1854	$3\ P_{3/2}$--$3^2S_{1/2}$
RbI	7947	2650	$5^2P_{1/2}$--$5^2S_{1/2}$
RbI	7800	2650	$5^2P_{3/2}$--$5^2S_{1/2}$
RbI	4216	2080	$6^2P_{1/2}$--$5^2S_{1/2}$
CsI	4555	2085	$7^2P_{3/2}$--$6^2S_{1/2}$
LiBr	6708	2040	$2^2P_{1/2}$--$2^2S_{1/2}$
NaBr	5896	2144	$3^2P_{1/2}$--$3^2S_{1/2}$
TlI	5351	2080	$7^2S_{1/2}$--$6^2P_{3/2}$
TlBr	5351	1915	$7^2S_{1/2}$--$6^2P_{3/2}$
TlCl	5351	1850	$7^2S_{1/2}$--$6^2P_{3/2}$

(RbI oven temperature is approximately 700°C)

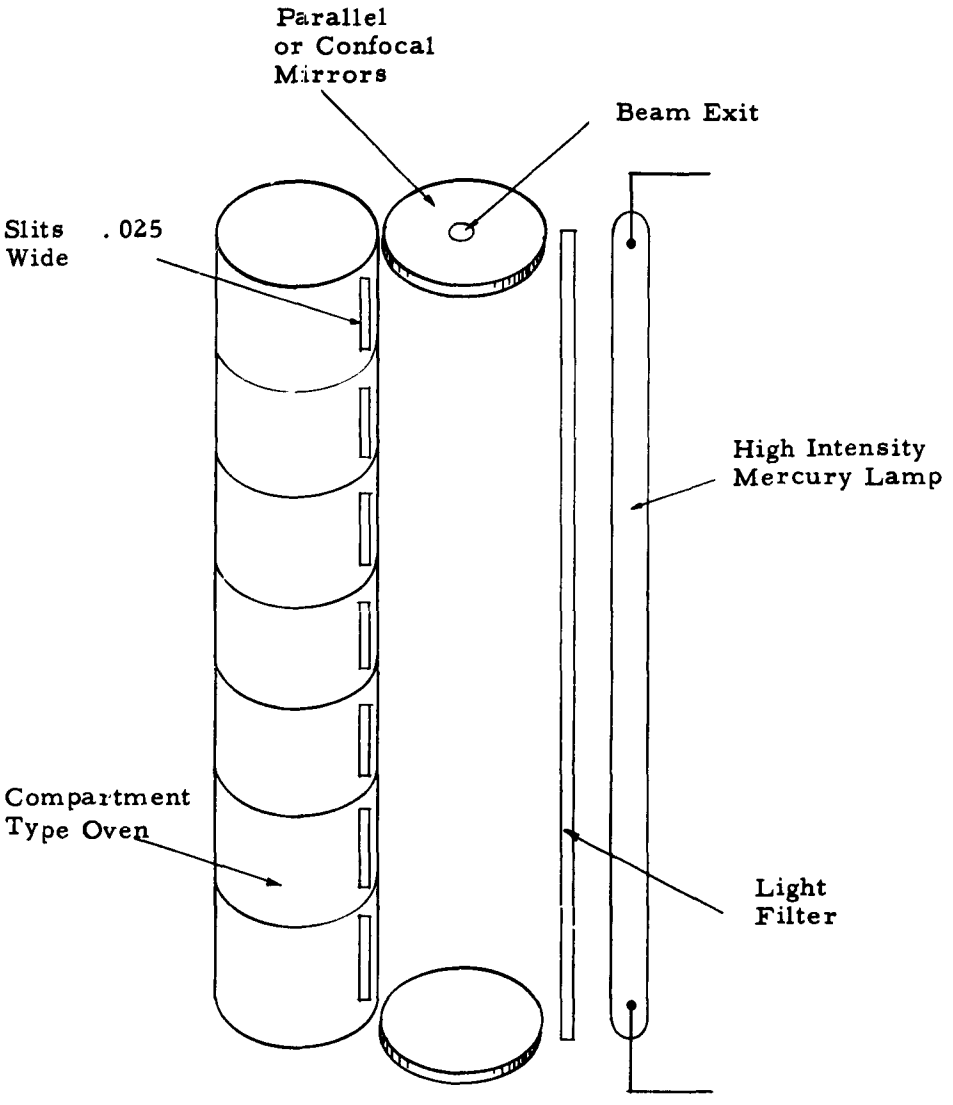

Figure 1. Schematic plan of molecular beam photodissociation type optical maser

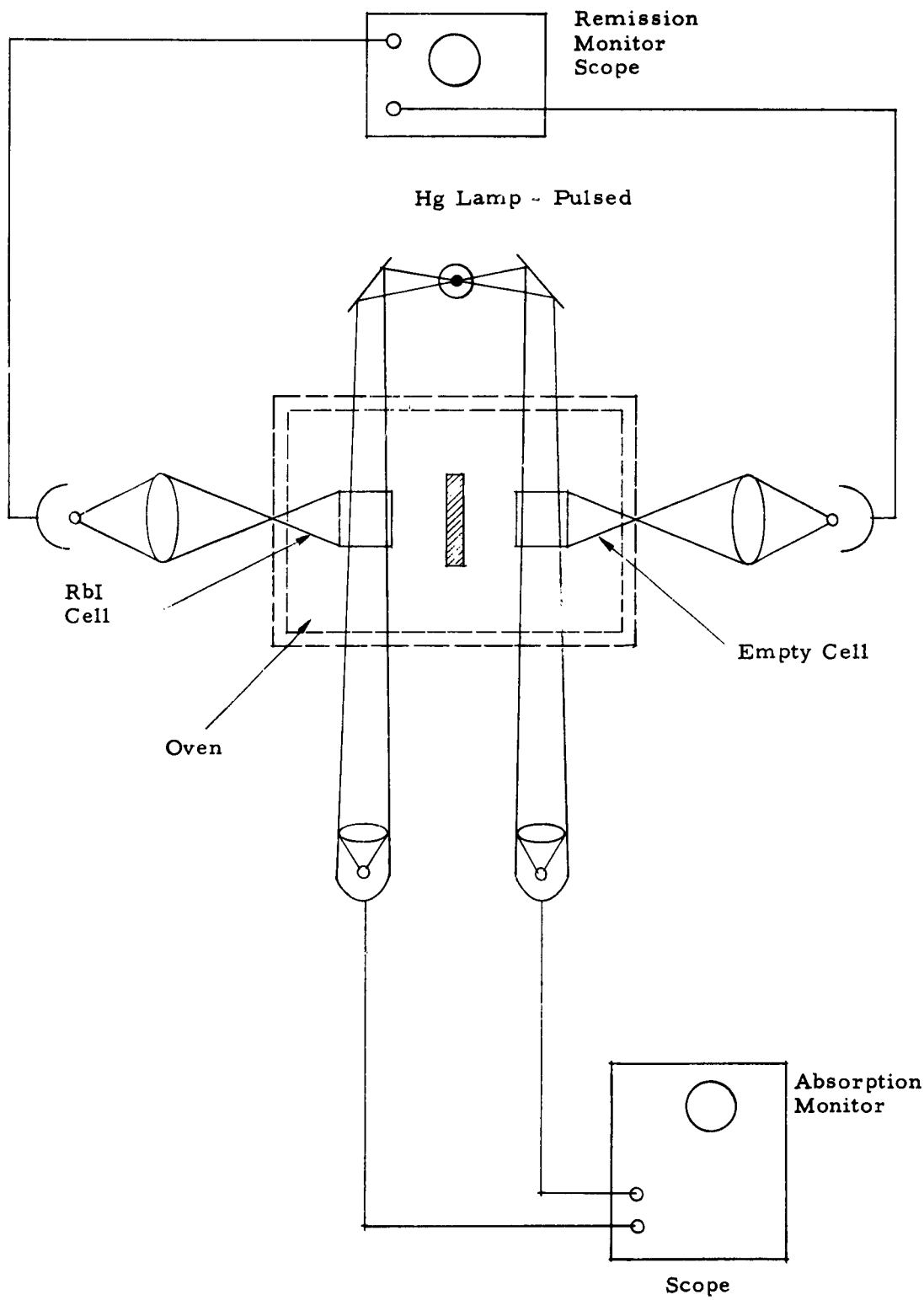

Figure 2. Experimental arrangement for measurement of the RbI photodissociation cross-section

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